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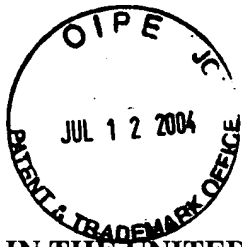
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**PATENT APPLICATION**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES**

In re application of

Docket No: Q65685

Takayuki YAMAMOTO, et al.

Appln. No.: 09/918,532

Group Art Unit: 1713

Confirmation No.: 3507

Examiner: Tatyana Zalukaeva

Filed: August 01, 2001

For: ACRYLIC PRESSURE-SENSITIVE ADHESIVE AND PROCESS FOR PRODUCING  
THE SAME

**SUBMISSION OF APPELLANT'S BRIEF ON APPEAL**

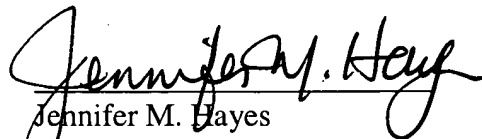
**MAIL STOP APPEAL BRIEF - PATENTS**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

Submitted herewith please find an original and two copies of Appellant's Brief on Appeal. Charge Deposit Account No. 19-4880 for the statutory fee of \$330.00. The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account. A duplicate copy of this paper is attached.

Respectfully submitted,

  
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WASHINGTON OFFICE

**23373**

CUSTOMER NUMBER

Date: July 12, 2004



**PATENT APPLICATION**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES**

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For: ACRYLIC PRESSURE-SENSITIVE ADHESIVE AND PROCESS FOR PRODUCING  
THE SAME

**APPELLANTS' BRIEF ON APPEAL UNDER 37 C.F.R. § 1.192**

**MAIL STOP APPEAL BRIEF - PATENTS**

Commissioner for Patents

P.O. Box 1450

Alexandria, VA 22313-1450

Sir:

In accordance with the provisions of 37 C.F.R. § 1.192, Appellant submits the following:

**I. REAL PARTY IN INTEREST**

The real party in interest is Nitto Denko Corporation of Osaka, Japan.

**II. RELATED APPEALS AND INTERFERENCES**

Appellants, Appellants' legal representative, and the Assignees of this application are not aware of any other appeals or interferences which will directly affect or be affected by or have a bearing on the Board's decision in the pending appeal.

**III. STATUS OF CLAIMS**

Claims 3-5 and 8-10 are canceled. This is an appeal from the Examiner's rejection of claims 1, 2, 6, 7, 11 and 12.

#### **IV. STATUS OF AMENDMENTS**

The Amendment under 37 C.F.R. § 1.116, filed on September 22, 2003, is entered in view of the Request for Continued Examination (RCE) and fee filed on September 22, 2003, as indicated in the Office Action dated November 25, 2003.

The Examiner did not specifically indicate whether the Amendment filed on April 26, 2004, correcting an error in the specification on page 13, line 19 would be entered. However, since the amendment to the specification is purely editorial to correct an obvious typographical error in the by changing "60□C" to "60°C", Appellants presume that the amendment has been entered for purposes of appeal.

#### **V. SUMMARY OF THE INVENTION**

The present invention relates to a process for producing an acrylic pressure-sensitive adhesive comprising continuously feeding a monomer, mainly comprising at least one alkyl (meth)acrylate, a radical polymerization initiator and carbon dioxide to a continuous reactor through a mixer and performing continuous bulk polymerization at a polymerization temperature of 50 to 180°C for a residence time of 0.5 to 200 minutes in a continuous reaction zone of the reactor. The acrylic pressure-sensitive adhesive comprises preferably 10% by weight or less, more preferably 6.5% by weight, based on the weight of the total monomers, of components having a molecular weight of 100,000 or less. Specification, page 4, lines 8-21.

Acrylic pressure-sensitive adhesives are widely used as a pressure-sensitive adhesive layer of an adhesive tape, an adhesive label or an adhesive sheet because of their excellent

pressure-sensitive adhesion characteristics (pressure adhesion and cohesion), weather resistance, oil resistance and the like. Specification, page 1, lines 13-18.

Commonly employed acrylic pressure-sensitive adhesives usually comprise a copolymer prepared from a monomer mixture comprising an alkyl (meth)acrylate which provides a tacky polymer having a relatively low glass transition temperature, such as n-butyl acrylate or 2-ethylhexyl acrylate, as a main monomer and, in addition, a monomer having a functional group for increasing crosslinking sites or for enhancing intermolecular forces thereby improving cohesiveness, such as acrylic acid, 2-hydroxyethyl (meth)acrylate or acrylamide, or a hard monomer providing a polymer having a relatively high glass transition temperature, such as styrene or vinyl acetate. Specification, page 1, line 19 to page 2, line 4.

These acrylic pressure-sensitive adhesives are generally produced by solution polymerization, suspension polymerization or emulsion polymerization. However, solution polymerization cannot get rid of the environmental pollution problem arising from use of a large quantity of an organic solvent. Also, isolation of the produced polymer requires an extra step and energy for vaporization of the organic solvent, which leads to an increased production cost. Specification, page 2, lines 4-11.

Suspension polymerization and emulsion polymerization are disadvantageous in that an emulsifying agent or a dispersing agent is incorporated into the resulting polymer, making it difficult to obtain a pure product. Isolation of the polymer involves the same problem as in solution polymerization. Moreover, these processes, carried out in a batch system, achieve poor

uniformity in polymerization conditions such as temperature and monomer concentration.

Specification, page 2, lines 11-19.

In view of the above, attempts to increase the polymerization conversion have resulted in a wider molecular weight distribution with a considerable proportion of low-molecular weight components. Existence of a large amount of low-molecular weight components can hinder pressure-sensitive adhesive properties and increase components that may be transferred to an adherend to cause contamination. Specification, page 2, lines 19-25.

Various proposals have been made for a continuous bulk polymerization technique in which a reactor comprising a single-screw or twin-screw extruder is used to carry out polymerization in a continuous manner. Bulk polymerization generally does not suffer from the above-described problems and is industrially advantageous. In a continuous process, since monomers are fed continuously, reduction in the monomer concentration, which occurs in the final stage of a batch-wise polymerization reaction, can be suppressed; thereby theoretically reducing production of low-molecular weight components. Further, in theory one skilled in the art can set the reaction conditions uniformly so that the polymerization temperature distribution can be made narrower. Specification, page 2 line 26 to page 3, line 11.

Thus, a continuous bulk polymerization process is expected to produce a polymer having a narrow molecular weight distribution in its ideal mode. In practice, however, depending on the monomers employed, it often occurs that the reaction proceeds abruptly to increase the viscosity of the system, which makes temperature control difficult, and a runaway reaction can result.

Therefore, molecular weight design of the polymer has been difficult. Additionally, in the past it has been difficult to completely eliminate a stagnant zone in a screw extruder used as a reaction apparatus, and gel or a deteriorated product may be formed, resulting in a failure to obtain a homogeneous polymer. Specification, page 3, lines 11-22.

The inventors of the present invention have conducted extensive investigations to solve the outstanding problems associated with conventional continuous bulk polymerization processes using alkyl acrylate monomers. They have found as a result that carbon dioxide used as a diluent serves to control the viscosity even with an abrupt reaction progress and thereby secures temperature control to avoid a runaway reaction. They have also found that carbon dioxide used as a diluent eliminates a stagnant zone in a reaction tube or a screw extruder thereby suppressing by-production of gel or a deteriorated product. Based on these findings the present invention was developed. Specification, page 3, line 24 to page 4, line 7.

The present invention is defined by two independent claims on appeal, namely claim 1 and claim 6. Claim 1 provides a process for producing an acrylic pressure-sensitive adhesive comprising:

- feeding a monomer solution comprising a mixture of an alkyl (meth)acrylate monomer and a radical polymerization initiator, and carbon dioxide to a joint block equipped with a line mixer;

- mixing the monomer solution and carbon dioxide in the joint mixer;

- feeding the resulting mixture to a continuous reactor; and

performing continuous bulk polymerization at a polymerization temperature of 50 to 180°C for a residence time of 0.5 to 60 minutes in a continuous reaction zone of said reactor, thereby obtaining an acrylic pressure-sensitive adhesive comprising 10% by weight or less, based on the weight of the total monomers, of components having a molecular weight of 100,000 or less.

Claim 6 provides a process for producing an acrylic pressure-sensitive adhesive comprising:

feeding a monomer solution comprising a mixture of an alkyl (meth)acrylate monomer and a radical polymerization initiator, and carbon dioxide to a joint block equipped with a line mixer;

mixing the monomer solution and carbon dioxide in the joint mixer;

feeding the resulting mixture to a continuous reactor; and

performing continuous bulk polymerization at a polymerization temperature of 50 to 100°C for a residence time of 60 to 200 minutes in a continuous reaction zone of said reactor, thereby obtaining an acrylic pressure-sensitive adhesive comprising 10% by weight or less, based on the weight of the total monomers, of components having a molecular weight of 100,000 or less.

## **VI. ISSUES**

1. Whether replacing the term “weight average molecular weight” with “molecular weight” can be considered as new matter to substantiate the rejection of claims 1, 2, 6, 7, 11 and 12 under 35 U.S.C. § 112, first paragraph, as allegedly containing subject matter not described in



the specification as originally filed and the corresponding objection to the amendment to the specification?

2. Whether the rejection of the claims as being indefinite under 35 U.S.C. § 112, 2<sup>nd</sup> paragraph should be reversed?

3. Whether the rejection of claims 1, 2, 6, 7, 11 and 12 under 35 U.S.C. § 103(a) as unpatentable over U.S. Patent 6,224,938 to Bamba et al should be reversed?

## **VII. GROUPING OF CLAIMS**

The claims stand or fall together for purposes of this appeal only.

## **VIII. ARGUMENTS**

**A. The term “molecular weight” has literal support in the original specification and does not constitute new matter.**

Claims 1, 2, 6, 7, 11 and 12 are rejected under 35 U.S.C. § 112, first paragraph, as allegedly containing subject matter which allegedly was not described in the specification in such a way as to reasonably convey to one skilled in the relevant art that the inventor(s), at the time the application was filed, had possession of the claimed invention. This rejection focuses on the amendment to the claims replacing the term “weight average molecular weight” with “molecular weight”.

Further, the amendment to the specification replacing the term “weight average molecular weight” with “molecular weight” is objected to under 35 U.S.C. § 132 as allegedly introducing new matter into the application.

The Examiner's position is that the introduction of the term "molecular weight" instead of the previously recited term "weight average molecular weight" is not supported by the original specification. Specifically, the Examiner asserts that the instant specification provides for a "weight average molecular weight" and does not mention or provide guidance for the term "molecular weight".

This rejection and objection are respectfully traversed. Appellants respectfully submit that the original specification provides literal support for the presently claimed invention and amendment to the specification. First, Appellants note that the original specification provides literal support for the term "molecular weight" as recited in the amended claims. For example, on page 6, lines 15-16 it states, "the resulting acrylic polymer has a high molecular weight with a reduced low molecular component content . . ." (emphasis added) and on page 8, lines 18-20, it states, "an acrylic pressure-sensitive adhesive having very less low molecular weight components can be obtained" (emphasis added). Therefore, contrary to the Examiner's statements, the original specification does expressly recite and provide support for the term "molecular weight" in describing the low molecular weight components of the acrylic polymer.

The claims and specification were amended to correct an obvious error in the description of "weight average" molecular weight in other locations, where the low molecular weight components -- as opposed to the entire polymer -- had been inadvertently characterized using the term "weight average molecular weight". It is well settled that an amendment to correct an obvious error is not considered as new matter where one skilled in the art would recognize the

existence of the error and the appropriate correction thereof. See *In re Oda*, 443 F.2d 1200, 1204 (CCPA 1971) (“That amendments may be made to patent applications for the purpose of curing defects, obvious to one skilled in the art, in the drawings or written descriptions of inventions, is so well settled that we deem it unnecessary to cite authorities in support thereof.”) See also, MPEP § 2163.06(II).

In this case, claims 1, 2, 6, 7, 11 and 12 and the specification were amended to correct an inadvertent error in referring to a “weight average molecular weight” with respect to the amount of the low molecular weight components, i.e., having a molecular weight 100,000 or less. These amendments are entirely consistent with the specification as filed, in view of the literal support in the original specification for the term “molecular weight” used in its proper context in relation to low molecular weight components of the acrylic polymer, as discussed above, and in view of the overall teachings in the disclosure as a whole.

It is clear that inadvertent mistakes were made in certain locations when the disclosure is considered overall. For instance, in the disclosure of the original specification, it is indicated in that the aim of the present invention is to narrow the molecular weight distribution and reduce the “*proportion of the amount of low-molecular [weight] components*” that can hinder pressure-sensitive adhesive properties and increase components that may be transferred to an adherend and cause contamination. See specification, page 2, lines 19-25 (emphasis added).

Further, it is clear from the examples in the original specification that the terms “weight average molecular weight” and “molecular weight” have different meanings. For example,

Example 1 refers to the bulk polymer having a weight average molecular weight of 1,020,000 (see page 10, lines 26-27), so it does not make any sense for the example to subsequently refer to the specific components thereof as having a weight average molecular weight of 100,000 or less (see page 11, lines 1-2). The skilled artisan would recognize that the components are discussed in terms of their actual molecular weight. This is consistent throughout the examples.

The term “molecular weight” is a term of art recognized by those of ordinary skill in the art to refer to relative molecular mass, which is obtained by adding together the atomic weights of the formula of the substance; or is determined by chemical or physical methods, such as lowering of freezing point, vapor pressure or vapor density. See Grant & Hackh's Chemical Dictionary, 5<sup>th</sup> Ed. 1987, page 375, previously submitted on April 26, 2004, which is attached for the Board's convenience as Attachment A. This definition of the term “molecular weight” is consistent with the use of the term in the original specification and as amended, as would be understood by one of ordinary skill in the art for the reasons discussed above.

Thus, “molecular weight” is an appropriate term to describe the physical properties of polymer components or building blocks of a polymer. In contrast, “weight average molecular weight” is a term used to describe the distribution of a polymer *per se* as discussed further below. See, e.g., U.S. Pat. Nos. 6,194,498 and 6,586,525 (previously submitted on April 26, 2004, and attached as Attachment B and C, respectively). Thus, one of ordinary skill in the art would have readily recognized the amendment to change the term “weight average molecular weight” to

“molecular weight” as the appropriate correction to correct the obvious error in the description of the polymer components as opposed to the polymer as a whole.

In addition, it is clear that the disclosure of 100,000 or less in the original specification refers to a range of molecular weights, such as a portion of Fig. 1-3 in Billmeyer, Textbook of Polymer Science 3<sup>rd</sup> Ed. (previously filed on September 22, 2003, and as Attachment D) starting from the left side of the x-axis (the molecular weight axis) and moving to a molecular weight of 100,000. The requirement in the claims of having 10% by weight or less of components having a molecular weight of 100,000 or less is similar to the EPA's Premanufacture Notification (PMN) requirement of a weight % below 500 molecular weight and a weight % below 1000 molecular weight. (See page 5 of the EPA's PMN Form attached as Attachment E.)

In view of the above, one skilled in the art, upon reading the original specification, would have understood that the term “weight average molecular weight,” was incorrect in the amended locations and that “molecular weight” is the proper term from the context of the specification.

Therefore, the presently claimed invention is adequately described in the specification as originally filed in view of the fact that (1) the term “molecular weight” in describing the polymer components is literally supported in the original specification as filed; (2) it is clear from the overall disclosure as originally filed that the specification as amended was intended to refer to “molecular weight” as opposed to “weight average” molecular weight in describing the polymer components; and (3) the term “weight average molecular weight” is an obvious error in the

amended locations and the term “molecular weight” is an appropriate correction as is readily recognized by one of ordinary skill in the art.

Accordingly, Appellants respectfully request that the rejection under 35 U.S.C. § 112, 1<sup>st</sup> paragraph, be reversed and the objection under 35 U.S.C. § 132 be withdrawn.

**B. The term “molecular weight” is art-recognized and is not indefinite; thus, the claimed invention is properly and sufficiently defined.**

Claims 1, 2, 6, 7, 11 and 12 are further rejected under 35 U.S.C. § 112, 2<sup>nd</sup> paragraph as allegedly being indefinite based on the amendment to the claims replacing the term “weight average molecular weight” with “molecular weight”. The Examiner states that the term “molecular weight” does not identify whether the recited molecular weight is determined based upon weight average, number average or viscosity average.

Appellants respectfully traverse the rejection for the following reasons.

As discussed above with respect to the rejection under 35 U.S.C. § 112, 1<sup>st</sup> paragraph, claims 1, 2, 6, 7, 11 and 12 and the objection to the specification under 35 U.S.C. § 132, the claims and specification were amended in this case to correct an inadvertent, obvious error in certain locations in referring to a “weight average molecular weight” with respect to the amount of the low molecular weight components, i.e., having a molecular weight 100,000 or less, which is consistent with and supported by the specification as filed. Based upon the plain language of the claims, it is readily understood by one of ordinary skill in the art that the term “weight average molecular weight” is incorrect and the term “molecular weight” is the appropriate correction to properly describe the low molecular weight components having a molecular weight

of 100,000 or less. The term “molecular weight” is art-recognized, and has a definite meaning. See Grant & Hackh's Chemical Dictionary at 375. Further, it is indicated in the original disclosure that *the aim of the present invention is to narrow the molecular weight distribution and reduce the “proportion of the amount of low-molecular [weight] components* that can hinder pressure-sensitive adhesive properties and increase components that may be transferred to an adherend and cause contamination. See specification, page 2, lines 19-25 (emphasis added). Therefore the definition of low molecular weight components having a molecular weight of 100,000 or less is consistent with the disclosure in the specification in that the number of these components having a molecular weight of less than 100,000 as opposed to a “weight average molecular weight” (which as shown on the GPC (gas permeation chromatography) chart below includes those having a molecular weight of 10,000,000.) Thus, when the claims are properly read in light of the specification, it is readily recognized by one of ordinary skill in the art that the claims are clear and definite in scope.

The term “molecular weight” refers to a measure of the size of a molecule and in the case of a specific molecule, its molecular weight is unequivocally determined (for example, methanol: 32, ethanol: 46, etc.). On the other hand, a polymer is an aggregate of molecules having different degrees of polymerization, and comprises smaller components having various molecular weights. Therefore, the “molecular weight” of a polymer component can clearly be expressed, but to express the molecular weight of a polymer as a whole, the average value of the molecular weights is employed, and “weight average” molecular weight is one expression for an average

value. Thus, the molecular weight of the “components having a molecular weight less than 100,000 or less” can be appropriately defined as “molecular weight,” because it refers to the amount of polymer components having a molecular weight of  $10^5$  or lower that are present with respect to the entire polymer.

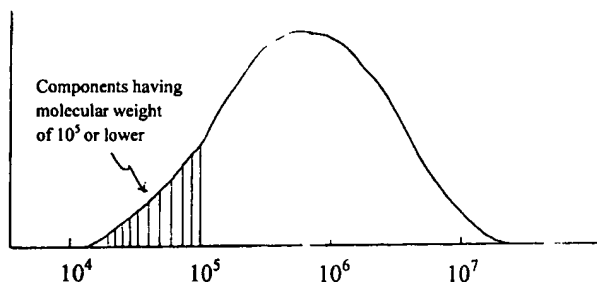
It is clear that the meaning intended throughout the specification and claims in the present application is the *amount of polymer components having a molecular weight of  $10^5$  or lower that are present with respect to the entire polymer* (see, GPC chart below), which is consistent with the disclosure and as is clear to one of ordinary skill in the art. As indicated in the original specification, the aim of the present invention is to narrow the molecular weight distribution and reduce the “*proportion of the amount of low-molecular [weight] components*” that can hinder pressure-sensitive adhesive properties and increase components that may be transferred to an adherend and cause contamination.

GPC can be used to measure the molecular weight distribution of a polymer. In the GPC chart below, the proportion of an area corresponding to the molecular weight of 100,000 or less can be calculated from the GPC chart (curve). In other words, the components having a molecular weight of 100,000 or less in the present invention include all those in the shaded area under the bell curve. In the present invention, Appellants define a molecular weight of 100,000 as the cut-off to be expressed as a specific molecular weight and maximum percentage present in the polymer as a whole. If the molecular weight is expressed as an “average” molecular weight, cut-off for the desired amount of low molecular weight components the would be unclear, since



the term “average molecular weight” would include polymers having a molecular weight higher than 100,000, by virtue of the definition of the term average.

GPC chart: weight average molecular weight of entire polymer =  $\sim 10^6$



Thus, the Examiner's position that the term “molecular weight” as presently recited in independent claims 1 and 6 should be “weight average”, “number average” or “viscosity average” is incorrect. When reading the claims, as amended, in view of the specification as a whole, one of ordinary skill in the art can readily ascertain the meaning and scope of the invention as claimed and therefore the claimed invention is adequately defined.

To further show that the term “molecular weight” by itself is art-recognized in the context used in Appellants' specification, submitted on April 26, 2004, Appellants refer to US Patent Nos. 6,194,498 (Attachment B), 6,444,772 (Attachment F), 6,586,525 (Attachment C) and 6,610,800 (Attachment G), which employ the term “molecular weight” as used in the present specification, and which establish a distinction between the terms “weight average molecular weight” and “molecular weight”.

For example, in US Patent No. 6,194,498, the term “weight average molecular weight” is discussed in relation to determining the polydispersity, or molecular weight distribution, of a

polyurethane which has a combination of “relatively high molecular weight polymers” and a substantial portion of “relatively low molecular weight polymers” (emphasis added). See col. 5, lines 42-48. Polydispersity is defined as the number derived from dividing the “weight average molecular weight” by the number average molecular weight at column 5, lines 55-57 and the low and high “molecular weight” portions are described as being based on “weight basis”. See col. 6, lines 3-6.

U.S. Patent No. 6,444,772, which was cited by the Examiner, is another example wherein the “molecular weight” of various polymers is disclosed in col. 2, lines 51-66 and in Tables I, II and III.

U.S. Patent No. 6,586,525 is an example of a patent where the term “molecular weight” is recited in the claims. See claims 1, 18 and 19. The invention relates to a composition that contains “not more than 2% of a component with molecular weight of 2,000 or lower and which has a ratio of weight average molecular weight (Mw) to number average molecular weight (Mn) . . .” emphasis added). See Abstract. It is also disclosed that the content of the “lower molecular weight component with weight average molecular weight (Mw) to number average molecular weight (Mn), that is, Mw/Mn were determined by means of gel permeation chromatograph (GPC), using polystyrene with a known molecular weight and narrow molecular weight distribution” (emphasis added). This issued patent is directly analogous to the use of terminology in the present application.

U.S. Pat. No. 6,610,800 is an example wherein it is disclosed that “molecular weight determination is deduced by using narrow molecular weight distribution standards” (emphasis added). U.S. Pat. No. 6,610,800 also discloses that the polydispersity index is typically measured by GPC (col. 7, lines 46-47); how equivalent polyethylene molecular weights can be determined (col. 7, lines 55-64); and how weight average molecular weight can be calculated using the weight fraction ( $w_i$ ) and molecular weight ( $M_i$ ) derived from the GPC column. (col. 7, line 65- col. 8, line 5).

In view of the above exemplary U.S. patents, Appellants submit that the term “molecular weight” is art-recognized and commonly used and understood by those of ordinary skill in the art. In addition, the disclosure of these patents regarding the terms “polydispersity”, “molecular weight distribution”, “molecular weight”, “weight average molecular weight” and “number average molecular weight” are consistent with the disclosure in Appellants’ specification as well as the arguments and evidence previously made of record.

In this regard we refer to the references previously submitted with the Amendment filed on September 22, 2003 (copies are attached for the Board’s convenience). For example, the x-axis in Fig. 1-3 on page 17 of Billmeyer, is simply labeled “Molecular Weight,” and the figure is described as showing the distribution of “molecular weights.” On page 41 in Stevens, Polymer Chemistry, 2<sup>nd</sup> Ed. (attached as Attachment H),  $M$  is the “molecular weight” of a species  $i$ . At the bottom of page 41, Stevens gives an example in which a polymer sample consists of 9 mol of molecular weight 30,000 and 5 mol of molecular weight 50,000, with these molecular weights

being used to calculate a number average molecular weight of 37,000. Page 5 of the Environmental Protection Agency's Premanufacture Notification (PMN) form requires the maximum weight % below "500 molecular weight" and "1000 molecular weight." All these examples are instances where the term "molecular weight" itself is used distinct from "weight average molecular weight." Thus, the term "molecular weight" by itself is well known in the art and not indefinite and when properly read in light of the specification, one of ordinary skill in the art would be able to readily ascertain the meaning and scope of the claims.

In the Advisory Action dated June 3, 2004, the Examiner cites *Ex parte Simpson*, 61 USPQ.2d. 1009 (Bd. Pat. App. & Int. 2001) in support of his position that the term "molecular weight" is indefinite. A copy of the Board decision is attached for the Board's convenience as Attachment I. In addition, the Examiner states that the arguments regarding other issued patents employing the term "molecular weight" is not persuasive because an Examiner is not bound by the decision of another Examiner with respect to patentability and the term "molecular weight" is used and cited in a different context in the issued patents provided.

Appellants note that *Ex parte Simpson*, referred to by the Examiner is an unpublished decision of the Board of Patent Appeals and Interferences and is non-precedential. Further, *Ex parte Simpson* can be distinguished from the present situation on the facts since the term "molecular weight" is literally supported and defined in the original specification as discussed in the Response filed on April 26, 2004 and herein, whereas the term "molecular weight" was not defined in the subject application of *Ex parte Simpson*. Further, *Ex parte Simpson* appears to

primarily relate to the probative value of a Declaration filed during prosecution of the subject application, which is not an issue in the present case.

With respect to the Examiner's statements regarding other issued patents employing the term "molecular weight" and the Examiner not being bound by another Examiner's decision regarding patentability, Appellants submit that *Ex parte Simpson* appears to favor Appellants' position. First, we note that the other issued patents were submitted with the response filed on April 26, 2004, "to further show that the term 'molecular weight' by itself is art-recognized in the context used in Appellants' specification" as indicated on page 10, lines 10-11 of the text of the Response. In other words, the other U.S. issued patents were submitted to indicate what the term would mean to a person of ordinary skill in the art which is the standard for determining definiteness of claim language under 35 U.S.C. § 112, 2<sup>nd</sup> paragraph. Further, the Board in *Ex parte Simpson* followed this same procedure in determining whether the term "molecular weight" was definite in that case by reviewing the prior art patents of record relied upon by the Examiner in support of the rejections under 35 U.S.C. § 103(a) and those mentioned in the specification of the application. Therefore, the other issued U.S. Patents are relevant in this context, i.e., the meaning of the term "molecular weight" as used in the present specification and claims and as would be understood by one of ordinary skill in the art.

Accordingly, Appellants respectfully submit that the rejection under 35 U.S.C. § 112, 2<sup>nd</sup> paragraph, should be reversed.

**C. Bamba does not teach or suggest all elements of the claimed invention and therefore does not render the claimed invention obvious.**

Claims 1, 2, 6 and 7 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Bamba et al (U.S. 6,224,938).

Appellants respectfully submit that the Examiner has not made a *prima facie* showing of obviousness since Bamba does not teach or suggest all elements of the claimed invention.

First, Bamba does not disclose or suggest the step of uniformly mixing a monomer mixture and an inert fluid and feeding the resulting mixture to a continuous reactor.

The claimed process is directed to a production process wherein a monomer mixture and an inert fluid are previously mixed uniformly by a line mixer, the resulting mixture is supplied to a continuous reactor, and polymerization is performed therein. Bamba does not contain any disclosure to teach this production process and therefore does not teach or suggest this element of the claimed invention.

Further, Bamba does not teach or suggest Appellants' claimed reaction parameters as admitted by the Examiner. As the Examiner points out in the Office Action dated November 25, 2003, Bamba does not disclose the claimed polymerization time or the features of the apparatuses for polymerization as recited in the present claims. It is the Examiner's position that the recitation of structural limitations of the apparatus for performing the recited steps of the process do not serve to limit the claim and that the time and temperature parameters recited in

the claim are result effective variables that are conventionally adjusted to achieve optimum performance.

Appellants respectfully submit that a variable must be recognized as contributing to a specific result before it can be acknowledged as *prima facie* obvious to determine the optimum or workable range of the variable. See MPEP §2144.05(II)(B) citing *In re Antonie*, 559 F.2d. 618 (CCPA 1977). In this case, Bamba does not teach suggest or even recognize the significance of the claimed reaction parameters in achieving the claimed pressures-sensitive adhesive of the present invention.

A characteristic feature of the present invention is the production of an acrylic pressure-sensitive adhesive comprising 10% by weight or less, based on the weight of the total monomers, of components having a molecular weight of 100,000 or less, which is accomplished by the recited process. Bamba is asserted to disclose that “the residual monomer and low molecular weight components, which have the possibility to lower the pressure-sensitive adhesive properties, can be evaporated off”. However, Bamba discloses that those low molecular weight components are evaporated off simultaneously with the evaporation of the inert fluid, and the molecular weight is considerably smaller (generally, several hundreds). By contrast, the polymer adhesive obtained in the present invention contains a small amount of components having a molecular weight of 100,000 or less. Thus the claimed invention differs considerably from Bamba.

Bamba does not disclose or suggest a production process wherein a monomer mixture and an inert fluid are previously mixed uniformly by a line mixer, the resulting mixture is supplied to a continuous reactor, and polymerization is performed therein. Specifically, Bamba does not disclose the process recited in claim 1, wherein the continuous bulk polymerization process is carried out at a polymerization temperature of 50 to 180°C for a residence time of 0.5 to 60 minutes in a continuous reaction zone of the reactor. Further, Bamba does not disclose the process recited in claim 6, wherein the continuous bulk polymerization process is carried out at a polymerization temperature of 50 to 100°C for a residence time of 60 to 200 minutes in a continuous reaction zone of the reactor. Therefore the Examiner has not set forth a *prima facie* showing that it would have been obvious to one of ordinary skill in the art to achieve the claimed invention based on the disclosure of Bamba.

Accordingly, it is respectfully requested that the rejection be reversed.

#### **IX. CONCLUSION**

The present Brief on Appeal is being filed in triplicate. Unless a check is submitted herewith for the fee required under 37 C.F.R. §1.192(a) and 1.17(c), please charge said fee to Deposit Account No. 19-4880.

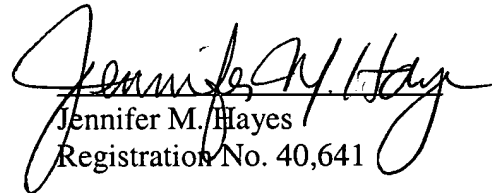


APPELLANTS' BRIEF ON APPEAL  
UNDER 37 C.F.R. § 1.192  
U.S. Appln. No.: 09/918,532

Attorney Docket No. Q65685

The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

Respectfully submitted,

  
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WASHINGTON OFFICE

**23373**

CUSTOMER NUMBER

Date: July 12, 2004

## APPENDIX

### CLAIMS 1, 2, 6, 7, 11 and 12 ON APPEAL:

1. (previously presented): A process of producing an acrylic pressure-sensitive adhesive comprising:

feeding a monomer solution comprising a mixture of an alkyl (meth)acrylate monomer and a radical polymerization initiator, and carbon dioxide to a joint block equipped with a line mixer;

mixing the monomer solution and carbon dioxide in the joint mixer;

feeding the resulting mixture to a continuous reactor; and

performing continuous bulk polymerization at a polymerization temperature of 50 to 180°C for a residence time of 0.5 to 60 minutes in a continuous reaction zone of said reactor, thereby obtaining an acrylic pressure-sensitive adhesive comprising 10% by weight or less, based on the weight of the total monomers, of components having a molecular weight of 100,000 or less.

2. (original) The process as claimed in claim 1, wherein said polymerization temperature is 65 to 130°C.

6. (previously presented): A process of producing an acrylic pressure-sensitive adhesive comprising:

feeding a monomer solution comprising a mixture of an alkyl (meth)acrylate monomer and a radical polymerization initiator, and carbon dioxide to a joint block equipped with a line mixer;

mixing the monomer solution and carbon dioxide in the joint mixer;

feeding the resulting mixture to a continuous reactor; and

performing continuous bulk polymerization at a polymerization temperature of 50 to 100°C for a residence time of 60 to 200 minutes in a continuous reaction zone of said reactor, thereby obtaining an acrylic pressure-sensitive adhesive comprising 10% by weight or less, based on the weight of the total monomers, of components having a molecular weight of 100,000 or less.

7. (original) The process as claimed in claim 6, wherein said polymerization temperature is 50 to 80°C.

11. (previously presented): The process as claimed in claim 1, wherein the acrylic pressure-sensitive adhesive comprises 6.5% by weight or less, based on the weight of the total monomers, of components having a molecular weight of 100,000 or less.

12. (previously presented): The process as claimed in claim 6, wherein the acrylic pressure-sensitive adhesive comprises 6.5% by weight or less, based on the weight of the total monomers, of components having a molecular weight of 100,000 or less.

# GRANT & HACKH'S CHEMICAL DICTIONARY

[*American, International, European and British Usage*]

*Containing the Words Generally Used in Chemistry,  
and Many of the Terms Used in the Related  
Sciences of Physics, Medicine, Engineering,  
Biology, Pharmacy, Astrophysics,  
Agriculture, Mineralogy, etc.*

*Based on Recent Scientific Literature*

FIFTH EDITION

*Completely Revised and Edited by*

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The previous edition of this book was *Hackh's Chemical Dictionary*,  
4th ed., published by McGraw-Hill in 1969. It was prepared by Dr.  
Julius Grant from a *Chemical Dictionary* compiled by Ingo W. D.  
Hackh. The current, or 5th, edition of this book was prepared by Dr.  
Roger L. Grant, whose father prepared the 4th edition.

*The editors for this book were Betty J. Sun and Susan Thomas,  
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supervisor was Teresa F. Leaden. It was set in Palatino  
by University Graphics, Inc.*

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concept may be visualized. (2) Mathematical description of process, financial, or economic interactions. **space lattice** ~ A group of wire nets and balls to show the arrangement of atoms in a crystal or molecule. Cf. *space lattice*.

**moderator** A substance used in a nuclear reactor (as, heavy water or graphite) to reduce the energy of the electrons.

**modification** (1) A slight alteration or change. (2) The conversion of cereal starch into a form in which it is readily acted on by enzymes, as in malting barley, or treating with heat.

**modified soda** A mixture of sodium carbonate and hydrogencarbonate; a cleanser.

**Moditen** Trademark for fluphenazine hydrochloride.

**Modrella** Trademark for a continuous-filament semimatt rayon yarn.

**modular** Describing a system of plant or apparatus construction or assembly, comprising a number of units which permit rapid erection and easy modification.

**modulus** The measure of a force or properties of mass or their effects. Often used as an abbreviation for Young's *m.*; as, high *m.* fiber. **bulk** ~ Compression modulus. The volumetric *m.* of elasticity, being the compressive force per unit cross section, divided by the change per unit volume.

**compression** ~ Bulk *m.* **shear** ~ Shear stress divided by shear angle. **wet** ~ See *high wet-modulus fiber* under *fiber*. **Young's** ~ Symbol: *E*; modulus of elasticity, *m.* of rigidity, longitudinal elasticity. The normal stress divided by the linear strain (relative elongation). Thus  $E = Fl/\pi r^2 s$ , where *s* is the elongation produced by a force *F* in a sample of length *l* and cross-sectional radius *r*; measured in MN/m<sup>2</sup>.

*m.* of elasticity, *m.* of rigidity Young's *modulus*.

**Mogadon** Trademark for nitrazepam.

**mohair** A long, lustrous textile fiber from the Angora goat.

**moho** Mohorovicic discontinuity. The boundary between the earth's inner mantle and the assorted surface rocks, 16-32 km beneath the surface.

**Mohr** M., Karl Friedrich (1806-1879) German chemist and physicist. **M. condenser** A modified Liebig condenser. **M. liter** Abbrev. g.w.a.; The space occupied by an amount of water at 17.5°C having an apparent weight in air (brass weights) of 1,000 g. 1,000 g.w.a. = 1,002 ml. **M. pipet** A small buret with tap, used as a pipet. **M. salt** (NH<sub>4</sub>)<sub>2</sub>Fe(SO<sub>4</sub>)<sub>2</sub>·6H<sub>2</sub>O. Ammonium iron(II) sulfate; a standard in volumetric analysis.

**Mohs** M., Friedrich (1773-1839) German mineralogist. **M. scale of hardness** The hardness of a mineral is gauged by its ability to scratch or be scratched by one of ten standard minerals:

1. Talc
2. Gypsum
3. Calcite
4. Fluorite
5. Apatite
6. Orthoclase
7. Quartz
8. Topaz
9. Corundum
10. Diamond

Each mineral is scratchable by all below it.

**Moissan** M., Ferdinand Frédéric Henri (1852-1907) French chemist, noted for the production of artificial diamonds and the isolation of fluorine. Nobel prize winner (1906). **M. furnace** A high-temperature electric furnace. **M. process**

The reduction of chromic oxide with carbon in an electric furnace lined with calcium chromite.

**moistness** The amount of liquid, generally water, held by a solid or gas. Cf. *wetting*, *absorption*.

**moisture** The wetness or dampness of a substance; the percentage of water contained in a substance.

**mol**\* Abbreviation for mole. **mol. wt.** Abbreviation for molecular weight.

**molal** Moles per weight, as in *m. solution*. **m. conductivity** Molar *conductance*\*. **m. solution** Concentration, q.v., expressed as molality; e.g., a 0.5 molal solution contains 0.5 mol/kg of solvent. Cf. *molar solution*. **m. volume** See *molar volume* under *volume*.

**molality**\* Symbol: *m*; concentration expressed in moles per kg of solvent. Cf. *molal solution*, *molar solution*.

**molar** (1)\* Before the name of an extensive quantity, means divided by the amount of substance; e.g., *molar volume*. Cf. *specific volume*. (2) Amount-of-substance concentration\*. Mole per volume. See *molar solution* below. **m. conductivity**

*M. conductance*\*. **m. latent heat** Molecular heat of vaporization. The quantity of heat (J or cal) required per mole to transform a substance from the liquid to the gaseous state.

**m. solution** A solution that contains one mole per liter of a substance. Thus a 1.0 M NaCl solution contains 58.5 g/liter.

**m. surface** The area of a sphere of one mole of a substance. **m. volume**\* See *molar volume* under *volume*. **m. weight** The molecular weight expressed in grams; a mole.

**molarity** (Amount-of-) substance concentration\*. Cf. *molar solution*, *molal solution*.

**molasses** Treacle. The uncrystallizable syrup obtained on boiling down raw cane or beet sugar solution (70% of sugars). Cf. *affination*.

**mold** Mould. (1) A receptacle in which a molten or liquid mass solidifies. (2) To shape or form. (3) The loose earth on the upper surface of cultivated soil. (4) A variety of fungoid growth, usually filamentous, which grows Hypomycetes, found on damp vegetable material; e.g., *Penicillium*.

**mole**\* Also *mol*\*. An SI base unit. The amount of substance of a system containing the same number (i.e., the Avogadro constant) of elementary entities (must specify which; e.g., atoms, molecules, ions, electrons, or other particles) as there are atoms in exactly 0.012 kg of carbon-12. Thus, 1 mole (gram molecule) of HgCl has a mass of 236.04 g.

**molecular** Pertaining to single molecules. Cf. *macromolecular chemistry*. **m. association** The state in which two or more molecules are held by coordinate bonds. **m. colloid** See *molecular colloid* under *colloid*. **m. combination** See *molecular combination* under *combination*. **m. compound** Double salt. **m. conductivity** Molar *conductance*\*. **m. conversion** See *rearrangement*. **m. depression** The lowering of the freezing point of a solution. See *Raoult's law*. **m. diagrams** Drawings to scale of a view of the *m. model*. They resemble structure symbols, but show the ionic and effective radii and the shape of the molecule. **m. diameter** The diameter of a molecule calculated from (1) Sutherland's equation, (2) van der Waals' equation, (3) the thermal conductivity, (4) the specific heat capacity at constant volume; e.g., in angstrom units:

	(1)	(2)	(3)
Hydrogen .....	2.40	2.34	2.32
Helium .....	1.90	2.65	2.30
Oxygen .....	2.98	2.92	
Nitrogen .....	3.18		

**m. dispersion** *M. rotation*. **m. elevation** The raising of the boiling point of a solution. See *Raoult's law*. **m. equation** See *chemical equation*. **m. field** See *molecular field* under *field*. **m. film** A monomolecular layer; as produced by adsorption. **m. flow** See *molecular flow* under *flow*. **m. formula** A combination of chemical symbols from which the *m. weight* of a substance is obtained by addition of the atomic weights.

m. conductivity  
tion, q.v.  
ion contains 0.5  
olume See molar

sed in moles per  
ion.

quantity, means  
olar volume. Cf.  
centration\*. Mole  
conductivity

heat of  
required per mole  
the gaseous state.  
le per liter of a  
ains 58.5 g/liter.  
e of a substance.

m. weight The  
le.  
tion\*. Cf. molar

up obtained on  
m (70% of sugars).

olten or liquid  
e loose earth on  
riety of fungoid  
Hypomycetes,  
nicillium.

ount of substance  
e., the Avogadro  
ify which; e.g.,  
particles) as there  
Thus, 1 mole

i.04 g.

Cf. macromolecular

ich two or more  
m. colloid See

ation See

m. compound

istance\*. m.

sion The lowering

ult's law. m.

re m. model. They

ionic and effective

diameter The

Sutherland's

the thermal

at constant volume;

The raising of the

v. m. equation See

field under field.

ced by adsorption.

i. formula A

nich the m. weight

he atomic weights

of the constituents. Cf. molecular formula. m. free path The mean free path of a molecule in a solution or gas, calculated from (1) Boltzmann's equation  $\eta(0.3592\rho c)$  or (2) Meyer's formula,  $\eta(0.3097\rho c)$ , where  $\eta$  is the viscosity,  $\rho$  the density of the medium, and  $c$  the molecular velocity. m. frequency The ratio  $v = k(T_s/MV^{2/3})^{1/2}$ , where  $v$  is the molecular frequency,  $T_s$  the melting point of the substance (in K),  $M$  the molecular weight,  $V$  the molar volume, and  $k$  a constant whose empirical value (Nernst) is  $3.08 \times 10^{12}$ . m. heat Specific heat capacity  $\times$  molecular weight. m. h. of vaporization See molar latent heat under latent. m. number (1) A number, analogous to the atomic number, obtained by arranging molecules according to their molecular frequencies. (2) The sum of the atomic numbers of the elements of a molecule; in a compound it is even, in a free radical it is odd. Cf. molecular combination. m. orbital See molecular orbital under orbital. m. rays A stream of molecules moving uniformly in one direction, obtained by the escape of vapor through an orifice into a vacuum, screening, and condensing the vapor on the wall of the vessel. m. rearrangement See rearrangement. m. rotation Specific rotation  $\times$  molecular weight. m. sieve A zeolite having an open-network structure, used to separate hydrocarbon and other mixtures by selective occlusion of one or more of the constituents; e.g., gmelinite adsorbs methane but not isoparaffins. m. solution A true solution, in which single molecules of the solute move in the solvent. m. s. volume The difference between the volume of a solution containing 1 mole/l of substance and that of 1 liter of solvent. m. velocity The mean velocity with which molecules move, proportional to the mean kinetic energy:  $19,300 \sqrt{(T/M)} \text{ cm/s}$ , where  $T$  is the thermodynamic temperature, and  $M$  the molecular weight. m. volume See parachor, volume. m. weight Symbol:  $M_r$ ; common name for relative molecular mass\*. The ratio of the average mass per formula unit of a substance to 1/12 of the mass of an atom of the nuclide  $^{12}\text{C}$ . Obtained by adding together the atomic weights indicated by the formula of the substance; or determined by chemical or physical methods; as, lowering of freezing point, vapor pressure, or vapor density. molecularity The number of reactant particles (as, atoms, molecules, or ions) taking part in a reaction, including any transition state.

molecule The chemical combination of 2 or more like or unlike atoms. The smallest quantity of matter that can exist in the free state and retain all its properties. In noble gases the m. is essentially monoatomic. Other gaseous m. usually consist of 2 atoms; as,  $\text{O}_2$ ,  $\text{Cl}_2$ . activated  $\sim$  A m. with one or more electrons at a higher energy level than their ground state. biatomic  $\sim$  Diatomic m. compound  $\sim$  A m. consisting of different atoms. elementary  $\sim$  A m. consisting of one type of atom. excited  $\sim$  Activated m. gram  $\sim$  Mole. homopolar  $\sim$  Homonuclear m. A diatomic m. composed of 2 similar atoms, e.g.,  $\text{H}_2$ . isosteric  $\sim$  One of a group of molecules having the same number of electrons, the same sum of atomic numbers, and, sometimes, the same molecular weight; as,  $\text{CO}_2$  and  $\text{N}_2\text{O}$ . nonpolar  $\sim$  See polar compound. oriented  $\sim$  A m. having directional properties. Cf. anisotropic. saturated  $\sim$  A m. in which all valencies are satisfied. symmetric top  $\sim$  A m. with an  $n$ -fold axis of symmetry, where  $n$  exceeds 2; e.g.,  $\text{CHCl}_3$ . spherical top  $\sim$  A m. having two axes of symmetry, e.g.,  $\text{CH}_4$ . tetraatomic  $\sim$  A m. having 4 atoms. tie  $\sim$  A m. in which parts of the same m. participate in more than 1 crystal lamella. triatomic  $\sim$  A m. having 3 atoms. unsaturated  $\sim$  A m. in which there are double or triple bonds, or both, between certain of the atoms.

molecules (constants) (1) Avogadro constant. The number of molecules per gram molecule (mole) is  $L = 6.022045 \times 10^{23}$ . (2) Loschmidt number. The number of molecules of a gas per mL at  $0^\circ\text{C}$  and 760 mm pressure is  $n = 2.687 \times 10^{19}$ . (3) Calculated constants:

Constant	$\text{H}_2$	$\text{O}_2$
Molecular weight	2.016	31.998
Velocity, m/s at $0^\circ\text{C}$	1,859	465
Mean free path, $\text{m} \times 10^{-9}$	965	560
Collisions, million/s	17,750	7,646
Diameter, $\text{m} \times 10^{-10}$	2.35	2.95
Mass, $\text{g} \times 10^{-25}$	46	736
Number per ml, $\times 10^{19}$	3.8	3.8

Typical molecular weights:

Insulin	5,634
Hemoglobin	65,000
Tobacco seed globulin	300,000
Hemocyanin	6,800,000
Tomato bushy stunt virus	10,000,000

molions The supposed negatively charged atomic groups of an ionized inert gas.

molybdaenum, molybdaen Early names for both native molybdenum sulfide and graphite, which were frequently confused.

molybdate\* Indicating molybdenum as the central atom(s) in an anion. Simple salts:  $\text{M}_2(\text{MoO}_4)$  or  $\text{M}_2\text{Mo}_2\text{O}_7$ , corresponding with the chromates and dichromates, respectively. Complex: polymolybdate(VI) acids and salts, analogous to the polytungstates, q.v.

molybdenic Molybdic.

molybdenite  $\text{MoS}_2$ . A white or green mineral.

molybdenous Molybdenum(II)\*,  $\text{Mo(II)}^*$ ,  $\text{Mo(2+)}^*$ . A salt of divalent molybdenum.

molybdenum\*  $\text{Mo} = 95.94$ . A heavy metal, at. no. 42, of the chromium group of the periodic table. Gray metal, d.10.2, m.2620, b.5560, insoluble in water or alkalis. It occurs in molybdenite, molybdite, wulfenite, and other rare minerals; and its presence in soil is important for the growth of grasses and vegetables. The metal is cast with difficulty and is used for crankshafts and connecting rods; as a resistor in heating devices and radios; as wire for vacuum tubes and contacts. Valency 2, 3, 4, 5, or 6; but the commoner compounds are derived from divalent  $[\text{Mo(II)}^*, \text{Mo(2+)}^*, \text{molybdenous}]$ , trivalent  $[\text{Mo(III)}^*, \text{Mo(3+)}^*, \text{molybdic}]$ , and hexavalent  $[\text{molybdic}, \text{molybdate(VI)}^*]$  molybdenum. m. blue  $\text{MoO}_2 \cdot 4\text{MoO}_3 \cdot \text{H}_2\text{O}$ . Heteropoly blue. A mixture of m. dioxide and trioxide. m. chlorides m. dichloride\*  $\text{MoCl}_2 = 166.8$ . Molybdenous chloride. Yellow, insoluble powder. m. trichloride\*  $\text{MoCl}_3 = 202.3$ . Molybdic chloride. Red needles. m. tetrachloride\*  $\text{MoCl}_4 = 237.8$ . Brown crystals. m. pentachloride\*  $\text{MoCl}_5 = 273.2$ . Black crystals, m.194. m. hexacarbonyl\*  $\text{Mo(CO)}_6 = 264.0$ . Colorless crystals. m. hydroxide (1)  $\text{Mo(OH)}_3 = 147.0$ . Black, insoluble powder. (2)  $\text{Mo(OH)}_5 = 181.0$ . Brown, insoluble powder. m. minerals Principal ores: molybdenite,  $\text{MoS}_2$ ; molybdite,  $\text{MoO}_3$ ; wulfenite,  $\text{PbMoO}_4$ . m. orange Pigments formed by the coprecipitation of lead molybdate, lead chromate, and lead sulfate in various proportions. m. oxides m. dioxide\*  $\text{MoO}_2$ . Blue prisms; a textile pigment. m. sesquioxide\*  $\text{Mo}_2\text{O}_3 = 239.9$ . Yellow to black mass, soluble in acid. m. trioxide\*

# TEXTBOOK OF POLYMER SCIENCE

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THIRD EDITION

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TABLE 1-4. Cohesive Energy Densities of Linear Polymers\*

Polymer	Repeat Unit	Cohesive Energy Density (J/cm <sup>3</sup> )
Polyethylene	—CH <sub>2</sub> CH <sub>2</sub> —	259
Polyisobutylene	—CH <sub>2</sub> C(CH <sub>3</sub> ) <sub>2</sub> —	272
Polyisoprene	—CH <sub>2</sub> C(CH <sub>3</sub> )=CHCH <sub>2</sub> —	280
Polystyrene	—CH <sub>2</sub> CH(C <sub>6</sub> H <sub>5</sub> )—	310
Poly(methyl methacrylate)	—CH <sub>2</sub> C(CH <sub>3</sub> )(COOCH <sub>3</sub> )—	347
Poly(vinyl acetate)	—CH <sub>2</sub> CH(OCOCH <sub>3</sub> )—	368
Poly(vinyl chloride)	—CH <sub>2</sub> CHCl—	381
Poly(ethylene terephthalate)	—CH <sub>2</sub> CH <sub>2</sub> OCOC <sub>6</sub> H <sub>4</sub> COO—	477
Poly(hexamethylene adipamide)	—NH(CH <sub>2</sub> ) <sub>6</sub> NHCO(CH <sub>2</sub> ) <sub>4</sub> CO—	774
Polyacrylonitrile	—CH <sub>2</sub> CHCN—	992

\*Walker (1952) and Small (1953).

properties typical of fibers, especially where molecular symmetry is favorable for crystallization. Chain stiffness or flexibility, referred to above, is largely determined by hindrance to free rotation about carbon-carbon single bonds in the polymer chain.

#### GENERAL REFERENCES

Ketelaar 1953; Cottrell 1958; Pauling 1960, 1964; Pimentel 1960; Chu 1967; Phillips 1970; Elias 1977, Part I.

#### D. MOLECULAR WEIGHT AND MOLECULAR-WEIGHT DISTRIBUTION

Perhaps the most important feature distinguishing polymers from low-molecular-weight species is the existence of a distribution of chain lengths and therefore degrees of polymerization and molecular weights in all known polymers (except possibly some biological macromolecules). This distribution can be illustrated by plotting the weight of polymer of a given molecular weight against the molecular weight, as in Fig. 1-3.

Because of the existence of the distribution in any finite sample of polymer, the experimental measurement of molecular weight can give only an average value. Several different averages are important. For example, some methods of molecular-weight measurement in effect count the number of molecules in a known mass of material. Through knowledge of Avogadro's number, this information leads to the

number-average  
number average  
probable m  
If the sar  
 $\sum_{i=1}^{\infty} N_i$ , and  
all the mol

and multip  
weight (m

Number  
range 10,0  
and others  
with typic  
After  $\bar{M}$   
absolute r  
defined as

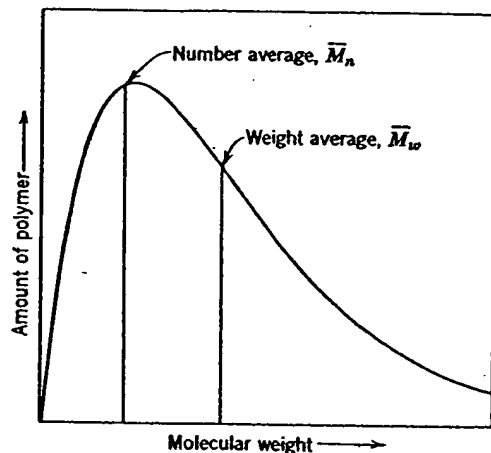


FIG. 1-3. Distribution of molecular weights in a typical polymer.

number-average molecular weight  $\bar{M}_n$  of the sample. For typical polymers the number average lies near the peak of the weight-distribution curve or the most probable molecular weight.

If the sample contains  $N_i$  molecules of the  $i$ th kind, for a total number of molecules  $\sum_{i=1}^{\infty} N_i$ , and each of the  $i$ th kind of molecule has a mass  $m_i$ , then the total mass of all the molecules is  $\sum_{i=1}^{\infty} N_i m_i$ . The number-average molecular mass is

$$\bar{m}_n = \frac{\sum_{i=1}^{\infty} m_i N_i}{\sum_{i=1}^{\infty} N_i} \quad (1-1)$$

and multiplication by Avogadro's number gives the number-average molecular weight (mole weight);

$$\bar{M}_n = \frac{\sum_{i=1}^{\infty} M_i N_i}{\sum_{i=1}^{\infty} N_i} \quad (1-2)$$

Number-average molecular weights of commercial polymers usually lie in the range 10,000–100,000, although some materials have values of  $\bar{M}_n$  10-fold higher, and others 10-fold lower. In most cases, however, the physical properties associated with typical high polymers are not well developed if  $\bar{M}_n$  is below about 10,000.

After  $\bar{M}_n$ , the next higher average molecular weight that can be measured by absolute methods is the weight-average molecular weight  $\bar{M}_w$ . This quantity is defined as

$$\bar{M}_w = \frac{\sum_{i=1}^{\infty} N_i M_i^2}{\sum_{i=1}^{\infty} N_i M_i} \quad (1-3)$$

TABLE 1-5. Typical Ranges of  $\bar{M}_w/\bar{M}_n$  in Synthetic Polymers\*

Polymer	Range
Hypothetical monodisperse polymer	1.000
Actual "monodisperse" "living" polymers	1.01-1.05
Addition polymer, termination by coupling	1.5
Addition polymer, termination by disproportionation, or condensation polymer	2.0
High conversion vinyl polymers	2-5
Polymers made with autoacceleration	5-10
Addition polymers prepared by coordination polymerization	8-30
Branched polymers	20-50

\*Billmeyer (1977).

It should be noted that each molecule contributes to  $\bar{M}_w$  in proportion to the square of its mass: A quantity proportional to the first power of  $M$  measures only concentration, and not molecular weight. In terms of concentrations  $c_i = N_i M_i$  and weight fractions  $w_i = c_i/c$ , where  $c = \sum_{i=1}^{\infty} c_i$ ,

$$\bar{M}_w = \frac{\sum_{i=1}^{\infty} c_i M_i}{c} = \sum_{i=1}^{\infty} w_i M_i \quad (1-4)$$

Unfortunately, there appears to be no simple analogy for  $\bar{M}_w$  akin to counting molecules to obtain  $\bar{M}_n$ .

Because heavier molecules contribute more to  $\bar{M}_w$  than light ones,  $\bar{M}_w$  is always greater than  $\bar{M}_n$ , except for a hypothetical monodisperse polymer. The value of  $\bar{M}_w$  is greatly influenced by the presence of high-molecular-weight species, just as  $\bar{M}_n$  is influenced by species at the low end of the molecular-weight distribution curve.

The quantity  $\bar{M}_w/\bar{M}_n$  is a useful measure of the breadth of the molecular-weight distribution curve and is the parameter most often quoted for describing this feature. The range of values of  $\bar{M}_w/\bar{M}_n$  in synthetic polymers is quite large, as illustrated in Table 1-5.

For some types of polymerization, the distribution of molecular weights (more often expressed as degrees of polymerization) can be calculated statistically; this topic is discussed in Chapter 3E. Experimental methods for measuring the molecular-weight averages defined above, among others, are the subject of Chapter 8.

#### GENERAL REFERENCES

Peebles 1971; Slade 1975; Billingham 1977.

#### BIBLIOGRAPHY

#### DISCUSSION

1. Define the of polym thermose
2. Discuss s how they lecular w
3. Consider  $M = 100$  adding th
  - a. 20 p
  - b. 20 p
 Calculate molecule
  - c. 20 p
  - d. 20 p
 Discuss molecule

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Press, Nev  
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J. Am. Ch  
Carothers 1931

## DISCUSSION QUESTIONS AND PROBLEMS

1. Define the following terms: polymer, monomer, repeat unit, network, degree of polymerization, homochain polymer, heterochain polymer, thermoplastic, thermosetting, configuration, conformation.
2. Discuss some of the properties that make polymers useful materials, and show how they result from unique features of polymer structure such as high molecular weight. (This topic is amplified in later chapters.)
3. Consider three hypothetical monodisperse polymers, with  $M = 10,000$ ,  $M = 100,000$ , and  $M = 1,000,000$ . For each, calculate  $\bar{M}_w$  and  $\bar{M}_n$  after adding the following to 100 parts by weight  $c_i$  of the polymer with  $M = 100,000$ :
  - a. 20 parts by weight of the polymer with  $M = 10,000$ .
  - b. 20 parts by weight of the polymer with  $M = 1,000,000$ .

Calculate  $\bar{M}_w$  and  $\bar{M}_n$  after adding the following to 100 parts by number (of molecules) of the polymer with  $M = 100,000$ :

- c. 20 parts by number of the polymer with  $M = 10,000$ .
- d. 20 parts by number of the polymer with  $M = 1,000,000$ .

Discuss the dependence of  $\bar{M}_w$  and  $\bar{M}_n$  on the presence of high- and low-molecular-weight material.

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# Part I. -- GENERAL INFORMATION -- Continued

## Section B -- CHEMICAL IDENTITY INFORMATION -- Continued

### 2. Polymers (For a definition of polymer, see the Instructions Manual.)

a. Indicate the number-average weight of the lowest molecular weight composition of the polymer you intend to manufacture. Indicate maximum weight percent of low molecular weight species (not including residual monomers, reactants, or solvents) below 500 and below 1,000 absolute molecular weight of that composition.

Describe the methods of measurement or the basis for your estimates: GPC ☐ Other ☐ : (Specify) \_\_\_\_\_

i) lowest number average molecular weight: \_\_\_\_\_

ii) maximum weight % below 500 molecular weight: \_\_\_\_\_

iii) maximum weight % below 1000 molecular weight: \_\_\_\_\_

☐ Mark (X) this box if you attach a continuation sheet.

b. You must make separate confidentiality claims for monomer or other reactant identity, composition information, and residual information. Mark (X) the "Confidential" box next to any item you claim as confidential.

- (1) -- Provide the specific chemical name and CAS Registry Number (if a number exists) of each monomer or other reactant used in the manufacture of the polymer.
- (2) -- Mark (X) this column if entry in column (1) is confidential.
- (3) -- Indicate the typical weight percent of each monomer or other reactant in the polymer.
- (4) -- Mark (X) the identity column if you want a monomer or other reactant used at two weight percent or less to be listed as part of the polymer description on the TSCA Chemical Substance Inventory.
- (5) -- Mark (X) this column if entries in columns (3) and (4) are confidential.
- (6) -- Indicate the maximum weight percent of each monomer or other reactant that may be present as a residual in the polymer as manufactured for commercial purposes.
- (7) -- Mark (X) this column if entry in column (6) is confidential.

Monomer or other reactant and CAS Registry Number (1)	Confidential (2)	Typical composition (3)	Identity Mark (X) (4)	Confidential (5)	Maximum residual (6)	Confidential (7)
		%			%	
		%			%	
		%			%	
		%			%	
		%			%	
		%			%	
		%			%	
		%			%	
		%			%	

☐ Mark (X) this box if you attach a continuation sheet.

c. Please identify which method you used to develop or obtain the specified chemical identity information reported in this notice. (check one).  
☐ Method 1 (CAS Inventory Expert Service - a copy of the identification report obtained from CAS Inventory Expert Service must be submitted as an attachment to this notice) ☐ Method 2 (other source)

d. The currently correct Chemical Abstracts (CA) name for the polymer that is consistent with TSCA Inventory listings for similar polymers.

e. Provide a correct representative or partial chemical structure diagram, as complete as can be known, if one can be reasonably ascertained.

☐ Mark (X) this box if you attach a continuation sheet.

# POLYMER CHEMISTRY

## An Introduction

SECOND EDITION

Malcolm P. Stevens  
University of Hartford

OXFORD UNIVERSITY PRESS  
1990

New York Oxford  
OXFORD UNIVERSITY PRESS  
1990

## 2

# Molecular weight and polymer solutions

### 2.1 Number average and weight average molecular weight

Molecular weight is an extremely important variable because it relates directly to a polymer's physical properties. In general, the higher the molecular weight, the tougher the polymer; however (as will be shown in the next chapter), too high a molecular weight can lead to processing difficulties. What defines an optimum molecular weight depends in large measure on the chemical structure of the polymer and the application for which it is intended. In this chapter we will be concerned with molecular weight definitions, methods of determining molecular weight, and methods of determining the distribution of molecular weights in a polymer sample. We will reserve our discussion of molecular structure for the following chapter. We will also be concerned with solution properties of polymers, because solubility is prerequisite to molecular weight determination.

Given that polymers are high-molecular-weight compounds, we must begin with the question, what do we *mean* by high molecular weight? Where does low molecular weight end and high begin? There is no simple answer, because what constitutes a "low" molecular weight for a sample of polyethylene, for example, might be ideal for a sample of polyamide. Furthermore, some polymers are deliberately prepared with low molecular weight (even as oligomers) to facilitate initial processing, molecular weight being increased at a subsequent processing stage. In general, however, we think of polymers as having molecular weights that run from the low thousands up to the millions, with optimum molecular weight depending on chemical structure and application. Vinyl polymers of any commercial import normally have molecular weights in the range  $10^5$  to  $10^6$ . Polymers having very polar functional groups, such as polyamides, may have molecular weights as low as 15,000 to 20,000.

To determine molecular weights of simple (nonpolymeric) compounds, we employ the familiar techniques of mass spectrometry, freezing-point depression (cryoscopy), boiling-point elevation (ebulliometry), and, where suitable functional groups are present, titration (for example, neutralization or saponification equivalents). Determining molecular weights of polymers, how-

ever, is considered polymerization chains to terminal average molecular weights provided by copy, ebulliometry, molecular-weight polymers with new techniques of polymer field because of the some exciting ne spectrometry in ever, in their traditional meth

Techniques for polymers include although titration used in some a determining mo this is not an absolute techniques c

Molecular weight of measurement properties (free: sure) give rise because the number. The total weight molecular species

where  $N$  and  $M$  of each species sample per mole

Suppose, for example, molecular weight

ever, is considerably more complex for two basic reasons. First, in any polymerization process, it is virtually impossible for all growing polymer chains to terminate at the same size; hence one must necessarily deal with *average* molecular weights. (Certain natural polymers having discrete molecular weights provide exceptions to this rule.) Second, the techniques of cryoscopy, ebulliometry, and titration are effective only with relatively low-molecular-weight polymers; more sophisticated methods must be used for polymers with molecular weights higher than about 40,000. The conventional techniques of mass spectrometry have not been used extensively in the polymer field beyond the characterization of polymer degradation products because of the requirements for volatilizable samples. Recent years have seen some exciting new developments in *field desorption* that have extended mass spectrometry into the macromolecular range. Such developments are, however, in their infancy and do not enjoy the routine applicability of the more traditional methods of molecular weight determination.

Techniques more commonly used for determining molecular weights of polymers include osmometry, light scattering, and ultracentrifugation, although titration (end-group analysis), cryoscopy, and ebulliometry are also used in some applications.<sup>1,2</sup> The most convenient method for routinely determining molecular weights involves measuring solution viscosities, but this is not an absolute method and can be used only in conjunction with one of the techniques of measuring absolute molecular weights.

Molecular weight values obtained depend in large measure on the method of measurement. Methods that depend on end-group analysis or colligative properties (freezing-point depression, boiling-point elevation, osmotic pressure) give rise to what is known as the *number average molecular weight* because the number of molecules of each weight in the sample are counted. The total weight of a polymer sample,  $w$ , is the sum of the weights of each molecular species present:

$$w = \sum_{i=1}^{\infty} w_i = \sum_{i=1}^{\infty} N_i M_i$$

where  $N$  and  $M$  are the number of moles and molecular weight, respectively, of each species  $i$ . Number average molecular weight,  $\bar{M}_n$ , is the weight of sample per mole:

$$\bar{M}_n = \frac{w}{\sum_{i=1}^{\infty} N_i} = \frac{\sum_{i=1}^{\infty} M_i N_i}{\sum_{i=1}^{\infty} N_i}$$

Suppose, for example, we have a polymer sample consisting of 9 mol of molecular weight 30,000 and 5 mol of molecular weight 50,000:

$$\bar{M}_n = \frac{(9 \times 30,000) + (5 \times 50,000)}{(9 + 5)} = 37,000$$



Suppose, instead, our sample consists of 9 g of molecular weight 30,000 and 5 g of molecular weight 50,000:

$$\bar{M}_n = \frac{(9 + 5)}{(9/30,000) + (5/50,000)} = 35,000$$

Light scattering and ultracentrifugation, on the other hand, are methods of determining molecular weight based on mass or polarizability of the species present. The greater the mass, the greater is the contribution to the measurement. In contrast to number average molecular weight (which is the summation of the *mole* fraction of each species times its molecular weight), these methods sum the *weight* fraction of each species times its molecular weight. The value thus obtained is called the *weight average molecular weight*,  $\bar{M}_w$ , and is expressed mathematically as

$$\bar{M}_w = \frac{\sum_{i=1}^{\infty} w_i M_i}{\sum_{i=1}^{\infty} w_i} = \frac{\sum_{i=1}^{\infty} N_i M_i^2}{\sum_{i=1}^{\infty} N_i M_i}$$

Consider the same two samples described previously. Nine mol of 30,000 molecular weight and 5 mol of 50,000 molecular weight:

$$\bar{M}_w = \frac{9(30,000)^2 + 5(50,000)^2}{9(30,000) + 5(50,000)} = 40,000$$

Substituting grams for moles:

$$\bar{M}_w = \frac{9(30,000) + 5(50,000)}{(9 + 5)} = 37,000$$

In each instance, we see that  $\bar{M}_w$  is greater than  $\bar{M}_n$ .

In measurements of colligative properties, each molecule contributes equally regardless of weight, whereas with light scattering, the larger molecules contribute more because they scatter light more effectively. It is for this reason that weight average molecular weights are always greater than number average molecular weights except, of course, when all molecules are of the same weight; then  $\bar{M}_w = \bar{M}_n$ . The narrower the molecular weight range, the closer are the values of  $\bar{M}_w$  and  $\bar{M}_n$ , and the ratio  $\bar{M}_w/\bar{M}_n$  may thus be used as an indication of the breadth of the molecular weight range in a polymer sample. This ratio is called the *polydispersity index*, and any system having a range of molecular weights is said to be *polydisperse*.

In our discussions of molecular weight measurement (Section 2.3), detailed descriptions of apparatus and derivations of working equations are not given. Students who wish to explore these areas in more detail are encouraged to consult the references provided. First, however, we will look briefly at *solubility* properties of polymers because the various methods of determining molecular weight or molecular weight distribution depend on solubility and polymer-solvent interactions.

## 2.2 Poly

Dissolving because of the Solution occurs the polymer student who solvent has formation.) dispersed into polymers dis periods of h Network po solvent. Even lack of solub type.

How does handbook, w polymers are modynamic p solubility. Ar

When a poly negative. The from increase magnitude of been propose to concentrati

where  $V_{\text{mix}}$  is (molecular w volume fracti terms  $\Delta E_1/V$   $(\Delta E/V)^{1/2}$  is )

The symbol  $\delta$  dissolve (nega be small. In Where  $\delta_1 = \delta_2$  solubility are solubility para densities.

**FULL TEXT OF CASES (USPQ2D)**

All Other Cases

**(Unpublished) Ex parte Simpson, 61 USPQ2d 1009 (BdPatApp&Int  
2001)**

**(Unpublished)  
61 USPQ2D 1009  
Ex parte Simpson**

**U.S. Patent and Trademark Office, Board of Patent Appeals and  
Interferences**

**No. 2000-0347  
Decided October 31, 2001**

**Unpublished Opinion**

(Unpublished)

**Headnotes**

**PATENTS**

**[1] Practice and procedure in Patent and Trademark Office —Prosecution —**

Page 1010

**Declarations/affidavits (§110.0913)**

**Patentability/Validity — Specification — Claim adequacy (§115.1109)****JUDICIAL PRACTICE AND PROCEDURE****Procedure — Evidence — Expert testimony (§410.3703)**

Patent examiner's decision to reject applicants' patent claims, directed to blend of linear low density polyethylene and high density polyethylene, for failure to adequately define term "molecular weight" is affirmed, even though applicants submitted declaration of inventor indicating that persons of ordinary skill in art would realize that "weight average molecular weight" was intended, since molecular weight can be measured in several different ways, since Manual of Patent Examining Procedure states that factual testimony is preferred over opinion testimony, since inventor's opinion is not supported by adequate documentation in record, and since applicants, therefore, have failed to establish that examiner erred in declining to accord inventor's testimony controlling weight; examiner's finding that inventor's opinion was "self-serving" relates to inventor's interest in outcome of case, which examiner was entitled to factor into analysis, and such finding should not be read as questioning inventor's good faith.

**Case History and Disposition**

Patent application of David M. Simpson and Alan M. Malakoff, serial no. 08/787,874, filed Jan. 23, 1997 (polyethylene blend). Applicants appeal from final rejection of claims for indefiniteness. Affirmed.

[Editor's Note: The Board of Patent Appeals and Interferences has indicated that this opinion is not binding precedent of the board.]

**Judge:**

Before Winters, and William F. Smith, administrative patent judges, and McKelvey, senior administrative patent judge.

**Footnotes**

1 Application for patent filed 23 January 1997. The real party in interest identified in the appeal brief is Union Carbide Chemicals & Plastics Technology Corporation. There is also a possibility that The Dow Chemical Company acquired an interest subsequent to the filing of the appeal brief.

**Opinion Text****Opinion By:**

McKelvey, S.J.

***Decision on appeal under 35 U.S.C. §134***

## ***A. Introduction***

The appeal is from a decision of a primary examiner rejecting claims 2, 6 and 7.

The appeal is about “molecular weight.” More particularly the appeal boils down to whether the examiner erred in declining to give controlling weight to declaration testimony of named inventor David M. Simpson. Because applicants have not established that the examiner erred in declining to give controlling weight to the declaration testimony of Dr. Simpson, we affirm.

## ***B. Findings of fact***

The record supports the following findings by at least a preponderance of the evidence.<sup>2</sup>

### ***The invention***

1. The claimed invention relates to a two-component blend of polyethylenes.
2. One polyethylene is a linear low density polyethylene (LLDPE).
3. The other can be a high density polyethylene (HDPE).
4. The prior art cited by the examiner will immediately confirm the fact that certain blends of LLDPE and HDPE are known. *See, e.g.*, Farley, U.S. Patent 5,530,065 (1996).
5. Applicants specify various parameters for each polyethylene component of the claimed blend, including density, melt index,<sup>3</sup>  $M_w/M_n$ ,<sup>4</sup> flow index.<sup>5</sup>
6. The claims require that the “molecular weight” of one polyethylene component be “in the range of about 50,000 to about 120,000” and that the “molecular weight” of the other polyethylene component be “in the range of about 190,000 to about 500,000”.
7. Applicants did not define in the specification what they mean by “molecular weight”.

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### ***The rejection***

8. Applicants'failure to say anything in the specification about what they mean by “molecular weight” has resulted in a controversy between applicants and the examiner as to the meaning of “molecular weight” in the claims.
9. In short, the examiner was unable to determine what applicants mean by “molecular weight”.
10. Accordingly, the examiner rejected the claims as being unpatentable under the second paragraph of 35 U.S.C. §112 as “indefinite.”
11. The examiner also made a plurality of prior art rejections.
12. As will become apparent, we do not reach the prior art rejections.

### ***Molecular weight of polyethylene***

13. The molecular weight of simple compounds, such as benzene, is not complicated. Basically, one adds up the atomic weight of each element of the compound and obtains the molecular weight.

14. In the case of polymers, including polyethylene, it is not so easy.

15. "Polymerization reactions, both synthetic and natural, lead to polymers with heterogeneous molecular weights, i.e., polymer chains with a different number of units."<sup>6</sup>

16. Accordingly, polyethylene, whether it be LLDPE or HDPE or any other form of polyethylene, generally consists of various polyethylene molecules, some of which are larger than others. Each polyethylene molecule therefore will have a different molecular weight.

17. The molecular weight of a particular polyethylene necessarily is an "average" molecular weight depending on the "average" molecular weight of each molecule in the polyethylene.<sup>7</sup>

18. What is "average molecular weight"<sup>8</sup> and how is it measured?

19. In general, those skilled in the art refer at least to "weight average molecular weight" and "number average molecular weight."

20. "The number-average value, corresponding to a measure of chain length of polymer chains, is called the *number-average molecular weight*. Physically, the number-average molecular weight can be measured by any technique that 'counts' the molecules. These techniques include vapor phase and membrane osmometry, freezing point lowering, boiling point elevation, and end-group analysis."<sup>9</sup> (Emphasis in original).

21. "In contrast to number average molecular weight (which is the summation of the *mole* fraction of each species times its molecular weight)," weight average molecular weight is the "sum [of] the *weight* fraction of each species times its molecular weight."<sup>10</sup>

22. Number average molecular weight is dependent on the number of molecules present in a particular molecular weight class, whereas weight average molecular weight is related to the weight of the molecules in a certain molecular weight class.<sup>11</sup>

23. We refer the reader to standard texts for a complete discussion of weight average molecular weight and number average molecular weight.<sup>12</sup>

24. "Molecular weight values obtained depend in large measure on the method of measurement."<sup>13</sup>

25. There are several methods for determining molecular weight. Typical methods include:<sup>14</sup>

a. Chromatographic techniques, to determine weight average molecular weight.<sup>15</sup>

b. Light scattering, to determine weight average molecular weight.

c. Membrane osmometry, to determine number average molecular weight.

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d. Vapor phase osmometry, to determine number average molecular weight.

- e. Electron and X-ray microscopy, to determine both number average and weight average molecular weight.
- f. Isopiestic method (isothermal distillation), to determine number average molecular weight.
- g. Ebulliometry (boiling point elevation), to determine number average molecular weight.
- h. Cryoscopy (melting point depression), to determine number average molecular weight.
- i. End-group analysis, to determine number average molecular weight.
- j. Osmodialysis, to determine number average molecular weight.
- k. Centrifugation, sometimes referred to as “ultracentrifugation,”<sup>17</sup> to determine weight average molecular weight.

26. Molecular weight distribution is also an important characteristic of polymers which can affect polymer properties. For example, a sample of polystyrene having a narrow molecular weight range will exhibit different properties from a sample having a broad range, even if the average molecular weights of the two samples are the same.<sup>18</sup>

27. Various methods exist for determining molecular weight distribution, the most widely used being gel permeation chromatography (GPC).<sup>19</sup>

### *Prosecution history*

28. Claim 4 of the application, as filed, contained a limitation that the “molecular weight” of the LLDPE had to be in the range of about 50,000 to about 120,000 and the “molecular weight” of the HDPE had to be in the range of about 190,000 to about 500,000.

29. In a first Office action (Paper 3), the examiner rejected claim 4 as being unpatentable under the second paragraph of 35 U.S.C. § 112 as indefinite.

30. The examiner observed (Paper 3, page 3) that applicants undoubtedly know that both LLDPE and HDPE are made up of a mixture of materials which have a range of molecular weights. The examiner acknowledged that in using the language “molecular weight” applicants presumably intended to refer to an “average” molecular weight. However, even if the word “average” appeared in the claim, the examiner would still find the claims indefinite “unless ‘weight’ or ‘number’ or some other basis of average molecular weight is specified” (Paper 3, page 3).

31. Applicants filed an amendment (Paper 4) in response to the first Office action, applicants cancelled claim 4 (as well as other claims) and added claims 6 and 7. Claims 6 and 7 referred to “weight average molecular weight.”

32. The examiner then entered a final rejection (Paper 5).

33. The examiner held that the “weight average” language of the “weight average molecular weight” limitation of claims 6 and 7 added “new matter” (Paper 5, page 2).

34. The examiner based his holding on a finding that the specification did not describe “molecular weight” as being “weight average molecular weight” (Paper 5, page 3).

35. In an amendment (Paper 6) after final rejection, applicants deleted the language “weight average” from the phrase “weight average molecular weight”(Paper 6, pages 1-2). According to remarks accompanying the amendment, the term “weight average”

was introduced through a misunderstanding of the examiner's comments in a previous Office letter [presumably the first Office action]. It is believed that a person of ordinary skill in the art would know that weight average molecular weight was meant by applicants. There are only two choices and this would be the common interpretation.

36. The examiner then entered a first “advisory” action (Paper 7), noting that the amendment would be entered upon filing of a notice of appeal.

37. The examiner observed (Paper 7, page 2) (emphasis added; bold in original):

Applicant's [sic—applicants'] argument that a person of ordinary skill in the art would know that weight average molecular weight was meant by applicants is not deemed to be persuasive because *it is not*

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supported by any evidence.[20] Further, applicant is [sic—applicants are] incorrect in stating that there are only “two choices”, as in addition to “weight average”and “number average” which are **both commonly used**, other molecular weights used to characterize polymers include at least “peak” or “peak average”, “viscosity average”and “z-average.”

38. Despite the fact that prosecution on the merits is closed after a final rejection, on 8 March 1999, applicants submitted a letter (Paper 9) accompanied by a first declaration of named inventor David M. Simpson.

39. The letter acknowledged that the examiner is correct that there are several expressions of molecular weight other than weight average and number average. According to counsel (Paper 9, page 1):

But to the best of my recollection, I have only seen these two used in claims, the most common by far being weight average. Number average is usually used as the denominator in the polydispersity ratio  $M_w/M_n$ , but it is used sometimes by itself.

40. The first declaration of Dr. Simpson reveals:

a. He received a B.S., an M.S. and a Ph.D. in Polymer Science from The Pennsylvania State University, the Ph.D. having been received in 1993.

b. Since 1994, he has been employed by the Union Carbide Corporation in its Research and Development Laboratory and has been primarily involved in the preparation and evaluation of polyolefin films.

c. He has reviewed the remarks of the examiner with respect to molecular weight, but respectfully disagrees.

d. He states:

In industry, within the Union Carbide Corporation, and in patents or other publications, when the term “molecular weight”is used without qualification to describe a polymer, a person of ordinary skill in the art would understand that weight average molecular weight was meant.

41. The examiner entered and considered the letter and first declaration and then filed a second “advisory” action (Paper 10).

42. The examiner declined to credit Dr. Simpson's declaration testimony. The examiner explained that the conclusion stated in the first declaration “is a self serving statement of an opinion unsupported by

any evidence.”<sup>21</sup> The examiner went on to observe (Paper 10, page 2):

[T]he Examiner wishes to point out that the claims also recite a melt index range for \* \* \* [one polyethylene] component and a flow index range for \* \* \* [the other polyethylene component]. If applicant can provide some evidence that these ranges could only be for the stated weight average molecular weights, then \* \* \* [the evidence] would be accepted as a basis for amended claims and specification reciting “weight average molecular weight.”

43. On 23 March 1999, the PTO received applicants' appeal brief (Paper 11). In the appeal brief (page 2), applicants called attention to Dr. Simpson's first declaration. Counsel for applicants repeated the argument which had been previously presented in the letter filed 8 March 1999 (Paper 9, page 1).

44. The examiner entered an Examiner's Answer (Paper 12) on 2 June 1999. Unknown to the examiner, on 16 April 1999, applicants had filed a second declaration of Dr. Simpson. As will become apparent below, it turns out that the second declaration had not been matched with the patent application file at the time the examiner entered his Examiner's Answer.

45. The examiner maintained his indefiniteness rejection based on the second paragraph of 35 U.S.C. §112 (Paper 12, page 4).

46. The examiner observed:

As Appellant [sic-applicants] undoubtedly knows [sic—know] both the LLDPE and the \* \* \* HDPE are a range of molecular weights and presumably an “average” molecular weight is intended although not stated [in the claim or specification]. However,

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it would also be indefinite unless “weight” or “number” or some other basis of average molecular weight is specified. One of ordinary skill in the art would recognize that both “weight average” and “number average” are commonly used to define average molecular weights. However, other bases of molecular weights are also used to characterize polymers including at least “peak” or “peak average”, “viscosity average” and “z-average”. One of ordinary skill in the art would also recognized that for the molecular weight distributions of the instantly claimed polymers, that the average molecular weights are significantly different from one another depending on the basis of measurement.

47. With respect to Dr. Simpson's first declaration, the examiner repeated (page 11) his prior observation that Dr. Simpson's opinion is a self serving statement unsupported by any evidence. Moreover, the examiner noted (page 12) that applicants acknowledge that the examiner was correct in noting that there are several expressions of molecular weight other than “weight average” and “number average” molecular weight and further acknowledge that number average molecular weight is sometimes used in claim language.

48. Applicants filed a reply brief (Paper 13) on 16 July 1999. Apparently, the second declaration of Dr. Simpson was still not matched with the patent application file as of the date of receipt of the reply brief.

49. In the reply brief, the following observation is made (Paper 13, page 1):

With regard to the 112 rejection, the examiner refers to appellant's [sic-applicants'] remarks with respect to molecular weight as “self serving” (sic). The dictionary defines self-serving as “serving one's own interests often in disregard of the truth ...”<sup>[22]</sup> The examiner is reminded that there is a duty of candor and good faith that is required both of appellants and the undersigned. We believe that we have met that duty, and that the



examiner is imposing an unfair burden on appellants, who are placed in the position where they have to make an empirical observation.

50. Ultimately, the second declaration of Dr. Simpson (Paper 15) reached the examiner.

51. The second declaration repeats the testimony set out in the first declaration. In addition, Dr. Simpson further testifies (Paper 15, page 2):

Further, it is well known by persons of ordinary skill in the art that melt indices and flow indices are inversely proportional to weight average molecular weights, and that is the case in the subject application. Thus, referring to present claim 6, the melt index of 0.5 to 30 grams per 10 minutes approximates the weight average molecular weight of 50,000 to 120,000 and the flow index of 0.4 to 11 grams per 10 minutes approximates the weight average molecular weight of 190,000 to 500,000.

The approximate correspondence between the melt and flow indices (melt flow properties) is proved by empirical means. The melt flow properties of a resin is [sic—are] determined by using an ASTM procedure, and the weight average molecular weight of the same resin is determined by using size exclusion chromatography (SEC). Plotting on a graph, the melt flow properties on the y-axis and the weight average molecular weight on the x-axis, the inversely proportional relationship between the two becomes apparent as well as the approximate correspondence.

52. In a third advisory action (Paper 16), the examiner entered and considered Dr. Simpson's second declaration.

53. The examiner agreed with Dr. Simpson that melt indices and flow indices are inversely proportional to weight average molecular weight (Paper 16, page 1). However, the examiner held that simply because the claims recite melt and flow indices does not mean that the molecular weights recited in the claims must therefore be weight average molecular weight. The examiner goes on to observe (pages 1-2):

[N]or is any *evidence*[23] presented to support a finding that the recited melt and flow index ranges correspond to the weight average

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molecular weights for the stated molecular weight ranges [in the claims].

54. The examiner continued to be of the opinion that applicants had “not provided any *evidence* that the molecular weights in question are ‘weight average molecular weights’”, citing §716.01(c) of the *Manual of Patent Examining Procedure* (MPEP).

### C. Discussion

Whether a claim is indefinite within the meaning of the second paragraph of 35 U.S.C. §112 is question of law to be resolved on the basis of underlying facts. *Credle v. Bond*, 25 F.3d 1566, 1576, 30 USPQ2d 1911, 1919(Fed. Cir. 1994). This appeal nevertheless essentially boils down to the weight to be given Dr. Simpson's declaration testimony. The examiner declined to give Dr. Simpson's declaration testimony controlling weight—despite the fact that the examiner found much in Dr. Simpson's declaration testimony with which he could agree. Applicants have not shown us why we should hold that the examiner erred and therefore we affirm. In short, we will not second-guess what is essentially a discretionary action taken by the examiner and which is supported by substantial evidence in this record.

The record establishes that, in this case, the term “molecular weight” needs to be defined. There is no definition in the specification. Hence, we are left to figure out what “molecular weight” would mean to a person

having ordinary skill in the art in the context of the claimed invention.

Dr. Simpson did not testify as to the level of skill in the art. However, the prior art relied upon by the examiner in support of his §103(a) rejections, and the prior art mentioned in the specification, tell us considerable about what one skilled in the art understands and what steps one skilled in the art takes to ensure that a skilled reader understands what is meant by molecular weight, at least in those cases where “molecular weight” is used in a claim.

Su, U.S. Patent 4,824,912 (Mobil Oil Corp.), as do applicants, describes a mixture of an LLDPE and HDPE, characterizing each a low or high molecular weight polymer. However, unlike applicants, the LLDPE and HDPE are claimed by reference to their respective melt indices. Examples of specific Exxon, Mobil and DuPont products are described in the specification. Hence, Su had no need to explicitly define the method by which molecular weight was to be determined.

Farley, U.S. Patent 5,530,065 (Exxon Chemical Patents, Inc.) describes and claims blends of polyolefins having certain molecular weight distributions (i.e.,  $M_w/M_n$ ). Farley, unlike applicants, explicitly describes a method for determining molecular weight distribution (col. 6, line 59 through col. 7, line 4).

Stehling, U.S. Patent 5,382,631 (Exxon Chemical Patents, Inc.) claims blends of polyolefins having “different average molecular weights”. See claim 1. However, unlike applicants, Stehling explicitly tells us that “average molecular weight” means “weight average molecular weight” (col. 5, lines 6-9).

Takahaushi, U.S. Patent 5,594,071 (Mitsui Petrochemical Industries, Ltd.) claims olefin compositions which are defined, *inter alia*, by their intrinsic viscosity. See part iii of Claim 1. As noted earlier, intrinsic viscosity is related to average molecular weight. See n.8, *supra*. Both the claims and the specification (col. 1, lines 63-65) of the Takahaushi patent describe how inherent viscosity is to be measured.

Hefner, U.S. Patent 5,194,532 (The Dow Chemical Co.), mentioned in applicants' specification (page 5), describes olefin copolymers having particular weight average molecular weights. Unlike applicants, however, Hefner explicitly states that “Weight average molecular weight by gel permeation chromatography (GPC) on a Water 150-C ALC/GPC using 3 Polymer Laboratories, Ltd. PL-gel 10 $\mu$ m mixed 300 x 7, 5 mm columns in series” (col. 18, Example 2 [Polymerization] and col. 20, footnote g after Table I).

Canich, U.S. Patent 5,227,440 (Exxon Chemical Patents, Inc.), unlike applicants, sets out precisely how a molecular weight determination is to be made (col. 16, line 49 through col. 17, line 6).

DeBoer, U.S. Patent 5,279,999 (Shell Oil Co.), unlike applicants, describes the use of catalysts to make polyolefins having “a number average molecular weight \* \* \* from about 200 to about 50,000” (col. 4, lines 64-67). Moreover, a method for determining number average molecular weight is explicitly described (col. 8, lines 51-53).

[1] The MPEP makes clear, “factual evidence is preferable to opinion testimony \* \* \*.” The MPEP also makes clear, “opinion” testimony is entitled to be considered, i.e. it is “admissible” in an ex parte proceeding. MPEP §716.01(c). The mere fact that opinion testimony is admissible (i.e., is entitled to be considered) does not *per se* mean it must be accorded controlling weight. In assessing the weight to be given expert testimony in an ex parte context, the

examiner may properly consider, among other things

- (1) The nature of the fact sought to be established.
- (2) The strength of any opposing evidence.
- (3) The interest of the expert in the outcome of the case.

(4) The presence or absence of factual support for the expert's opinion. There is no magic formula for resolving credibility and weight to be given declaration testimony. Rather, both credibility and weight determinations are made on a case-by-case basis through the exercise of informed judgment. Our experience reveals that in general item (4) tends to be significant. Although examiners and members of this board have scientific backgrounds, unless an "expert" states the underlying basis for an opinion, it may be difficult to accord the opinion significant weight.<sup>24</sup>

The examiner, in effect, was requesting Dr. Simpson to establish the underlying basis for his opinion that when someone says molecular weight they mean weight average molecular weight unless otherwise indicated. The examiner manifestly was justified in making the request. The prior art of record, including prior art mentioned in the specification, reveals that others have explicitly identified the nature of the "molecular weight" to which they make reference. Applicants acknowledge the correctness of the examiner's finding that molecular weight can be measured in different ways. Moreover, the examiner seems to have invited additional proof that polyethylenes having a certain melt index or a certain melt flow necessarily would have weight average molecular weights. Applicants apparently have declined the examiner's invitation.

The examiner could properly decline to accord controlling weight to Dr. Simpson's opinion that in industry "molecular weight" means weight average molecular weight unless otherwise specified. The record contains no convincing documentary evidence which would serve as an underlying basis for Dr. Simpson's opinion.

The examiner also could properly decline to accord controlling weight to any practice within Union Carbide Corporation. There is no evidence that one skilled in the art would, or could, be aware of Union Carbide practice. *Cf. Boyd v. Quigg*, 7 USPQ2d 1904 (D.D.C. 1988) (court rejected testimony based on "in-house" knowledge, holding that one skilled in the art might not be aware of "in-house" knowledge).

The examiner agreed with Dr. Simpson that there is an inverse relationship between (1) melt and flow indices and (2) weight average molecular weight. As correctly noted by the examiner, however, there is no convincing documentary evidence in the record to support a finding that the melt and flow indices recited in the claims correspond to weight average molecular weights for the molecular weights recited in the claims.

The examiner also could properly decline to accord Dr. Simpson's "approximate correspondence" discussion any weight. No actual example of a correspondence between melt or flow indices and weight average molecular weight was presented to the examiner. Moreover, if Dr. Simpson had established a numerical relationship between the melt and flow indices and the molecular weight recited in the claims, then we would want to inquire into why molecular weight limitations would be needed at all in the claims.

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Counsel for applicants also favored the examiner with his opinion on the matter. However, an opinion of counsel cannot take the place of evidence in the record. *See* cases cited in n.20, *supra*.

We have not overlooked, and we are sure the examiner did not overlook, the argument of counsel in the reply brief with respect to the duty of candor and good faith. *See* Finding 49 and 37 CFR §1.56. The examiner's finding that Dr. Simpson's opinion is "self-serving" should have been construed by counsel as an indication by the

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examiner that Dr. Simpson has an interest in the outcome of the case. An interest in the outcome of a case is a matter a decision-maker can take into account in any legal or administrative proceeding and is specifically set out in the MPEP as a factor to be considered. All will agree that it is not controlling. But, it is not irrelevant as applicants seem to assert. The examiner is no way questioned, nor do we, Dr. Simpson's good faith. The difficulty is not in Dr. Simpson's stated opinion. Rather, his opinion was not adequately supported by appropriate documentary support. Contrary to counsel's argument, the examiner did not impose an unfair burden on applicants. Rather, consistent with the provisions of MPEP §716.01(c) and jurisprudence generally applicable to legal and administrative proceedings in this country, the examiner considered, but declined to give controlling weight to, Dr. Simpson's declaration testimony.

Applicants have failed to establish that the examiner erred in declining to accord Dr. Simpson's testimony controlling weight. Since applicants have not shown that the examiner erred in determining that the term "molecular weight" in claims 2, 6 and 7 is indefinite within the meaning of the second paragraph of 35 U.S.C. §112, we affirm.

#### *D. Prior art rejections*

The examiner has entered and maintained numerous alternative rejections based on the prior art. We decline to reach those rejections given that the language of the claims has been held to be indefinite. *Cf. In re Wilson*, 424 F.2d 1382, 1385, 165 USPQ 494, 496 (CCPA 1970) (all words in a claim must be considered; if a word has no meaning, the subject matter does not become obvious — the claim becomes indefinite); *In re Steele*, 305 F.2d 859, 134 USPQ 292 (CCPA 1962) (claimed subject matter does not become obvious due to indefiniteness of language of a claim).

#### *E. Decision*

Upon consideration of the appeal, and for the reasons given, it is

ORDERED that the decision of the examiner rejecting claims 2, 6, and 7 under the second paragraph of 35 U.S.C. §112 is *affirmed*.

FURTHER ORDERED that we decline to reach the merits of the rejections based on the prior art.

FURTHER ORDERED that no time period for taking any subsequent action in connection with this appeal may be extended under 37 CFR §1.136(a).

**AFFIRMED**

#### **Footnotes**

2 To the extent these findings of fact discuss legal issues, they may be treated as conclusions of law.

3 Determined under ASTM D-1238, Condition E (specification, page 3).

4 Mw/Mn is known as the "polydispersity index." *See Stevens, Polymer Chemistry*, Oxford University Press, page 42 (2d ed. 1990).

5 Determined under ASTM D-1238, Condition F (specification, page 3).

6 Carraher, "Polymer Chemistry," Marcel Dekker, Inc., page 81 (4th ed. 1996).

7 Stevens, *supra*, at 41.

8 We will note that the intrinsic viscosity of a polymer in solution, like the viscosity of a melt, is related to the average molecular weight of a polymer. Carraher, *supra* at 67. Applicants do not purport to define molecular weight by way of intrinsic viscosity.

9 Carraher, *supra* at 84.

10 Stevens, *supra*, at 42.

11 Encyclopedia of Chemical Technology, John Wiley & Sons, Volume 18, page 207 (3d ed. 1982).

12 Carraher, "Polymer Chemistry," Marcel Dekker, Inc., pages 64-73, 77, 81-106 (4th ed. 1996); Stevens, "Polymer Chemistry," Oxford University Press, pages 40-42 and 59-66 (2d ed. 1990) and Encyclopedia of Chemical Technology, John Wiley & Sons, Volume 18, pages 207-221 (3d ed. 1982).

13 Stevens, *supra*, at 41.

14 Carraher, *supra*, at 88. *See also* Stevens, *supra*, at 41-42 and Encyclopedia of Chemical Technology, *supra*, at 208.

15 Encyclopedia of Chemical Technology, *supra*, at 208 and 211.

16 Encyclopedia of Chemical Technology, *supra*, at 210.

17 Encyclopedia of Chemical Technology, *supra* at 212.

18 Stevens, *supra*, at 59.

19 Stevens, *supra*, at 61.

20 The examiner's observation is manifestly correct given that an argument of counsel cannot take the place of evidence in the record. *See, e.g., Estee Lauder, Inc. v. L'Oreal, S.A.*, 129 F.3d 588, 595, 44 USPQ2d 1610, 1615 (Fed. Cir. 1997) (argument of counsel does not take the place of evidence); *In re Pearson*, 494 F.2d 1399, 1405, 181 USPQ 641, 646 (CCPA 1974) (same).

21 We construe the examiner's language "any evidence" to mean any supporting or underlying documentary evidence. The testimony of Dr. Simpson is, of course, "evidence."

22 Counsel did not favor the board with a citation to the precise dictionary from which the definition is said to have been taken.

23 Again, we construe the examiner's language "any evidence" to mean any supporting or underlying documentary evidence, including reports of actual laboratory testing. As noted earlier, the testimony of Dr. Simpson is "evidence."

24 In this respect, we will call attention to practice in patent interference cases before the board. In interference cases, a party is required to have an expert state the underlying basis of any opinion. *See* §42 of the Trial Section's STANDING ORDER:

§42. *Affidavits of expert witnesses*

Affidavits expressing an opinion of an expert must disclose the underlying facts or data upon which the opinion is based. *See* Fed. R. Evid. 705 and 37 CFR §§1.639(b) and 1.671(b).

Opinions expressed without disclosing the underlying facts or data may be given little, or no, weight. *See Rohm and Haas Co. v. Brotech Corp.*, 127 F.3d 1089, 1092, 44 USPQ2d 1459, 1462 (Fed. Cir. 1997) (nothing in the Federal Rules of Evidence or Federal Circuit jurisprudence requires the fact finder to credit the unsupported assertions of an expert witness).

**- End of Case -**